REPORT DOCUMENTATION PAGE

Form Approved
OMB No. 0704-0188

1. AGENCY USE ONLY *(Leave blank)*

2. REPORT DATE
3/26/98

3. REPORT TYPE AND DATES COVERED
Final Technical Report 8/92 - 12/97

4. TITLE AND SUBTITLE
Engineered Semiconductor Nanostructures for Enhanced Nonlinear Optical Properties in the Ingrared

5. FUNDING NUMBERS
G# N0014-92-J-4063

6. AUTHOR(S)
Philippe M. Fauchet

7. PERFORMING ORGANIZATION NAMES(S) AND ADDRESS(ES)
University of Rochester
Department of Electrical Engineering
Rochester, NY 14627-0231

8. PERFORMING ORGANIZATION REPORT NUMBER

9. SPONSORING / MONITORING AGENCY NAMES(S) AND ADDRESS(ES)
Office of Research & Project Administration
518 Hylan Building
Rochester, NY 14627

10. SPONSORING / MONITORING AGENCY REPORT NUMBER

11. SUPPLEMENTARY NOTES

a. DISTRIBUTION / AVAILABILITY STATEMENT
Available to the public

19980417 144

13. ABSTRACT *(Maximum 200 words)*

GaAs and InGaAs superlattices have been grown by molecular beam epitaxy. Doping was done either in the quantum wells or throughout the superlattices. The quantum wells were first characterized by conventional methods such as PL, FTIR, or Raman spectroscopy. The free electron laser (or another subpicosecond laser source tunable in the mid-infrared) was used to perform second harmonic generation (SHG) measurements and differential transmission measurements using the pump-probe configuration. The results were analyzed using the simplest realistic models available in the literature as well as the most advanced numerical models through collaborations. All measurements were performed in the 3-5 m spectrum region. We have measured for the first time (1) SHG enhancement on resonance with inter-subband transitions in the valence band, (2) the hot hole relaxation in p-type quantum wells, and (3) the hot electron relaxation in n-type quantum wells.

14. SUBJECT TERMS Free Electron Laser, nonlinear and time-resolved optical properties of semiconductor nanostructures, pump-probe, second harmonic generation, hot electrons, hot holes.

15. NUMBER OF PAGES
6

16. PRICE CODE

17. SECURITY CLASSIFICATION OF REPORT	18. SECURITY CLASSIFICATION OF THIS PAGE	19. SECURITY CLASSIFICATION OF ABSTRACT	20. LIMITATION OF ABSTRACT
unclassified	unclassified	unclassified	

FINAL REPORT

OFFICE OF NAVAL RESEARCH

Grant # N00014-92-J-4063

Engineered Semiconductor Nanostructures for Enhanced Nonlinear Optical Properties in the Infrared

PROFESSOR PHILIPPE M. FAUCHET

Department of Electrical Engineering
&
Laboratory for Laser Energetics
University of Rochester
Rochester NY 14627
Tel: (716)-275-1487

Date: March 11, 1998

Grant #: N00014-92-J-4063

PRINCIPAL INVESTIGATOR: Dr. Philippe M. Fauchet

INSTITUTION: University of Rochester

GRANT TITLE: Engineered semiconductor nanostructures for enhanced nonlinear optical properties in the infrared

AWARD PERIOD: 8-92 to 12-97

OBJECTIVE: To use the Free Electron Laser to perform a study of the nonlinear and time-resolved optical properties of engineered semiconductor nanostructures, including pump-probe experiments involving intersubband resonances in the valence band or in the conduction band of quantum wells; to compare the experimental results to theoretical predictions; to assess the feasibility of mid-infrared optical devices using these effects.

Dr. Philippe M. Fauchet/University of Rochester April 1998

SUMMARY:

Asymmetric, p-doped GaAs/AlGaAs quantum wells are grown and characterized by conventional techniques. The second harmonic generation (SHG) from these quantum wells is measured as a function of wavelength, polarization and power. After removing the contribution from the substrate, the amplitude and phase of the SHG signal is plotted versus wavelength and compared to the predictions of a first-principles theory.

Experiments have been conducted on several multiple quantum well AlGaAs/GaAs structures with the following characteristics: the doping is p-type ($2\ 10^{18}\ cm^{-3}$), the well width is 6 or 7 monolayers, the well is asymmetric (m GaAs monolayers and n $Al_{.5}Ga_{.5}As$ monolayers, with M=6, n-1, m=5, n=1; m=5, n=2; m=4, n=2), the barrier is 4 nm of AlAs, the total multiple quantum well thickness is between 1 and 2 µm, and the substrate is GaAs. FTIR spectra showed weak peaks in the region from λ <4 µm to > 5 µm, depending on the sample, and in broad agreement with simple bandstructure calculations. The band-to-band PL spectra of the samples were consistent with a bandgap in the red part of the visible spectrum, due to the large quantization energy in both valence band and conduction band. Summarizing the key results from the SHG experiments, we found, for the first time, a clear enhancement of the signal compared to the bulk GaAs reference near resonance. The magnitude of the enhancement was 10 at resonance and decreased with increasing detuning. Unlike what is known of SHG at 10.6 µm in n-type GaAs quantum wells, the symmetry properties of the SHG were identical in the quantum wells and in the bulk. A theoretical investigation of SHG in our structures performed by Prof. Jaros and his group (university of Newcastle, UK) showed excellent qualitative and acceptable quantitative agreement with our results.

Our results are the first ever such measurements involving the valence band. This research program is important because it gives information that allows precise control of the effective mass and wavefunction by changing the quantum well size, composition and strain. In addition, it may lead to improved optical and optoelectronic devices in a region of the spectrum where devices such as photodetectors or modulators are not as performant as in the visible or near infrared. These devices in turn can be used to detect and manipulate the FEL beam, for example in medical studies using the FEL.

The transient bleaching is measured in p-doped InGaAs/AlGaAs quantum wells as a function of temperature, laser wavelength and intensity. The results are modeled using analytical calculations involving confined phonons.

We have performed the first subpicosecond time-resolved bleaching on p-doped, strained InGaAs/AlGaAs quantum wells, using the pump-probe configuration. The signal recovery occurs on a 1psec time scale and depends on temperature and laser intensity, but not laser wavelength. We have interpreted these results as a direct measurement of the initial hot hole-phonon scattering time. The results of these studies have led to several publications.

Dr. Philippe M. Fauchet/University of Rochester April 1998

We have performed extensive measurements and preliminary calculations of the carrier dynamics in doped InGaAs square quantum wells. We have performed pump-probe measurements with a resonant pump (tuned to the n=1 to n=2 intersubband maximum) and either a resonant probe or a near bandgap probe (tuned near transition from the n=1 valence band level to the n=1 conduction band level). The excited electrons relax on a time scale of 1-2 psec, but the relaxation is complex (i.e., non-exponential) especially when a large fraction of the cold electrons are promoted to a higher subband by the pump pulse. This is the first observation of the complete excited carrier "history" following intersubband pumping.

Our results are the most extensive study of the carrier dynamics in the mid-infrared. We expect that it will help engineers and device physicists to build faster and more efficient optical and optoelectronic devices in a region of the spectrum where devices such as photodetectors or modulators are not as efficient as in the visible or near-infrared. These devices in turn can be used to detect and manipulate the FEL beam, for example in medical studies using the FEL. They should also find uses in applications such as pollution detection, high speed communications from satellite to satellite, manipulation of chemical reactions, and countermeasures.

PUBLICATIONS:

1. "Picosecond Nonlinear Optics in Semiconductor Quantum Wells with the SCA Free Electron Laser," Xu, Z. et.al.(1993) Free-Electron Laser Spectroscopy in Biology, Medicine and Materials Science, H.A. Schwettman editor, SPIE Vol. 1854, p. 69.

2. "Pump-probe and Second Harmonic Generation Measurements in Semiconductors," Fauchet, P.M. (1993). Presented at the October 1993 ONR Free-Electron Laser Program on Materials Symposium, Vanderbilt University, Nashville TN.

3. "Nonlinear Optical Properties of Ultranarrow p-type GaAs Quantum Wells," Xu, Z.(1994) et.al.Mat. Res. Soc. Symp. Proc. 326, 573.

4. "Second Harmonic Generation in p-type Quantum Wells," Quantum Well Intersubband Transition Physics and Devices, Xu, Z. et.al. (1994) H.C. Liu et. Al. Editors, Kluwer Academic Publishers (Dordrecht), Series E: Applied Sciences, Vol. 270, pp. 457-466.

5. "Second Harmonic Generation in p-type Quantum Wells,"Xu, Z. et.al. (1994) Quantum Well and Superlattice Physics V, G.H. Dohler and E.S. Koteles editors, SPIE Vol. 2139, p. 321.

6. "Second-Harmonic Generation In p-Type Asymmetric GaAs-$Al_xGa_{1-x}As$-AlAs Superlattices Due To Excitations Between Valence Minibands,: Shaw, M.J., Jaros, M. Xu, Z. Fauchet, P.M., Rella, C.W., Richman, B.A., Schwettman, H.A. and Wicks, G.W., Phys. Rev. B 50, 18,395-18,419 (1994).

7. "Hole Relaxation in p-Type $In_xGa_{1-x}As/Al_yGa_{y-1}As$ Quantum Wells Observed By ultrafast Midinfrared Spectroscopy," Xu, Z. Fauchet, P.M., Rella, C.W., Richamn, B.A., Schwettman, H.A., and Wicks, G.W., Phys. Rev. B 51, 10631-10634 (1995).

8. "Second Harmonic Generation near 4 μm In p-Type Asymmetric GaAs/AlGaAs/AlAs Quantum Wells," Xu, Z., Fauchet, P.M., Rella, C.W., Richamn, B.A., Schwettman, H.A., and Wicks, G.W., Solid State Commun. 93, 903-907 (1995).

9. "Mid-Infrared Femtosecond Spectroscopy Of Intersubband Hot Hole Relaxation In Quantum Wells," P. M. Fauchet, G. W. Wicks, Ju. V. Vandyshev, Z. Xu, C. W. Rella, and H. A. Schwettman, in *Ultrafast Phenomena X*, edited by P.F. Barbara, J.G. Fujimoto, W.H. Knox, and W. Zinth (Springer-Verlag, Berlin, 1996), pp. 398-399.

10. "Ultrafast Excitation And Deexcitation Of Local Vibrational Modes In A Solid Matrix: The Si-H Bond In Amorphous Silicon," Z. Xu, J. V. Vandyshev, P. M. Fauchet, C. W. Rella, H. A. Schwettman, and C. C. Tsai, in *Ultrafast Phenomena X*, edited by P.F. Barbara, J.G. Fujimoto, W.H. Knox, and W. Zinth (Springer-Verlag, Berlin, 1996), pp. 410-411.

11. "Ultrafast Excitation And De-Excitation Of The Si-H Stretching Mode In a-Si:H," Z. Xu, P. M. Fauchet, C. W. Rella, H. A. Schwettman, and C. C. Tsai, J. Non-Cryst. Solids 198-200, 11-14 (1996).

12. "Temperature Dependence Of The Intersubband Hole Relaxation In p-Type Quantum Wells," Z. Xu, G. W. Wicks, C. W. Rella, H. A. Schwettman, and P. M. Fauchet, in *Hot Carriers in Semiconductors*, edited by K. Hess, J.-P. Leburton and U. Ravaioli (Plenum, New York, 1996), pp 65-68.

13. "Mid-Infrared Femtosecond Spectroscopy of Intersubband Hot Carrier Relaxation in Quantum Wells," P. M. Fauchet, Ju. V. Vandyshev, J. M. Russell, T. A. Gardiner, Z. Xu, and G. W. Wicks, abstract presented at the International Conference on Semiconductor Microstructures and Superlattices, Liege, Belgium, July 1996.

14. "Mid-Infrared Femtosecond Spectroscopy of Intersubband Carrier Relaxation in Quantum Wells," Ju. V. Vandyshev, J. M. Russell, P. M. Fauchet, and G. W. Wicks, presented at the Optical Society of America Annual Meeting, Rochester, NY, October 1996.

15. "Femtosecond Near- And Mid- Infrared Spectroscopy Of Hot Electrons In Doped InGaAs Quantum Wells," T. A. Gardiner, Ju. V. Vandyshev, G. W. Wicks, and P. M. Fauchet, presented at the Quantum Electronics and Laser Science Conference, Baltimore, MD, May 1997.

16. "Femtosecond Infrared Spectroscopy Of Hot Electrons In An $In_{0.53}Ga_{0.47}As/In_{0.52}Al_{0.48}As$ Multiple Quantum Well Structure," T. A. Gardiner, Ju. V. Vandyshev, G. W. Wicks, and P. M. Fauchet, in *Ultrafast Electronics and Optoelectronics*, edited by M. Nuss and J. Bowers, OSA Trends in Optics and Photonics Series Vol. 13 (Optical Society of America, Washington, DC, 1997), pp 280-283.

CONFERENCE PRESENTATIONS:

Second Workshop on Optical Properties of Mesoscopic Semiconductor Structures, Snowbird UT, April 1993

1."Nonlinear Optics in Ultranarrow p-type GaAs Quantum Wells"

Conference on lasers and Electro-Optics, Baltimore MA, Mary 1993

1. "Second Harmonic Generation in ultranarrow p-type Stepped GaAs Quantum Wells"

ONR Free-Electron Laser Program on Materials Symposium, Vanderbilt University, Nashville TN, October 1993)

1. "Pump-probe and Second Harmonic Generation Measurements in Semiconductors"

IEEE Lasers and Electro-Optics Annual Meeting, San Francisco, October 1995

1. Harmonic Generation and Time-Resolved Spectroscopy in p-type Quantum Wells, Philippe M. Fauchet and Zhiwei Xu (Invited)

International Conference on Amorphous Semiconductors, Kobe, Japan, September 1995

1. Ultrafast Excitation and Deexcitation of the Si-H Stretching Mode in a-Si:H, Z. Xu, P.M. Fauchet, C.W. Rella, H.A. Schwettman, and C.C. Tsai

Free Electron Laser User Workshop, New York, August 1995

1. Picosecond Intersubband Hole Relaxation in p-type Quantum Wells, Z. Xu, P.M. Fauchet, C. W. Rella, H.A. Schwettman, and G.W. Wicks
2. Excitation and Deexcitation of the Si-H Stretching Mode in a-Si:H with Picosecond Free Electron Laser Pulses, Zhwei Xu, Philippe M. Fauchet, Chris W. Rella, H. Alan Schwettman, and Chuang Chuang Tsai

International Conference on Hot Carriers in Semiconductors, Chicago, July 1995

1. Temperature Dependence of the Intersubband Hole Relaxation in p-Type Quantum Wells, Z. Xu, G.W. Wicks, C.W. Rella, H.A. Schwettman, and P.M. Fauchet

Quantum Electronics and Laser Science Conference, Baltimore, May 1995

1. Direct Mid-Infrared Spectroscopic Measurements of Hole Relaxation in InGaAs/AlGaAs Quantum Wells, Z. Xu and P.M. Fauchet, C.W. Rella, B.A. Richman, H.A. Schwettman and G.W. Wicks

Quantum Optoelectronics Conference, Dana Point, March 1995

1. Hole Relaxation in p-type InGaAs/AlGaAs Quantum Wells Observed by Ultrafast Mid-Infrared Spectroscopy, Z. Xu and P.M. Fauchet, C.W. Rella, B.A. Richman, H.A. Schwettman, and G.W. Wicks